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(NASA-CR-150476) ANALYSIS OF THE DEPLETION OF A STORED AEROSOL IN LOW GRAVITY Final Report, 5 Jan. 1976 - 30 Jun. 1977 (Nevada Univ.) 34 p HC A03/MF A01 CSCI 07D G3/25

N78-12169

Unclas 53649

ANALYSIS OF THE DEPLETION OF A STORED AEROSOL IN LOW GRAVITY

CONTRACT No. NAS8-31780

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FINAL REPORT FOR PERIOD JANUARY 5, 1976 - JUNE 30, 1977

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DEC 1977
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NASA STI FACILITY

DATE OF REPORT: NOVEMBER 23, 1977

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ABSTRACT

The depletion of an aerosol similar to those to be used in the ACPL, stored in a container with linear dimensions of order 50 cm, has been studied in 1-g and in low gravity. Models were developed for sedimentation, coagulation and diffusional losses to the walls. The overall depletion caused by these three mechanisms is predicted to be of order 5 to 8% per hour in terrestrial conditions, which agrees with laboratory experience. Applying the models to a low gravity situation indicates that there, only coagulation will be significant. (Gravity influences diffusional losses because of convection currents caused by random temperature gradients). For the types of aerosol studied, the rate of depletion of particles for which $S_c < 10^{-2}$ (1 percent), should be somewhat less than $(10^{-3}N)$ percent per hour, where N is the concentration per cm^3.

1. INTRODUCTION

In experimental work with cloud condensation nuclei (CCN), samples are frequently stored in Mylar bags which have internally aluminized walls, in order to avoid the increased particle losses which can be caused by electric fields within a container with non-conducting walls. Experience in terrestrial laboratories shows that the loss rates are rather variable, but are usually of order 5-10% per hour for CCN with critical supersaturations (S_c) ranging up to about 10^{-2} (1%) stored in containers with linear dimensions of order 50 cm.

The question addressed in this report is how would terrestrial experience with stored aerosols compare with what would be expected to occur in the ACPL in low gravity (taken here as $10^{-4}g = 10^{-1}cm\ sec^{-2}$). In order to make such a comparison, it is obviously necessary to: (1) develop models which show how gravity affects the various mechanisms of aerosol depletion, and (2) check the overall validity of these models by applying them to the terrestrial situation and comparing the total losses predicted as a result of all depletion mechanisms with the laboratory experience quoted above.

It is obvious that particle loss as a result of fall-out depends on gravity in a very direct way, while the modification of the aerosol as a result of coagulation (which may cause either an increase or a decrease in the CCN count) is independent of gravity. Nevertheless, in order to meet objective (2) above, it

is necessary to make an estimate of the change in the CCN count which would be expected to result from coagulation among the particles in a laboratory aerosol for which overall loss rates of order 5-10% per hour are experienced. For the same reason, although loss due to fall-out would not be expected to be significant in low gravity, the losses occurring in 1-g need to be estimated.

The estimates derived below for losses due to both coagulation and sedimentation in 1-g are of order 1% per hour, which is small compared with that found experimentally. The chief result of this investigation is that most of the depletion found in 1-g is due to diffusional losses of particles to the walls in the presence of convection currents caused by ambient temperature gradients, which tend to continuously flush the walls with undepleted air. According to the model developed, the depletion rate due to convection-accentuated diffusional losses in 1-g is comparable with that observed. In low gravity (10^{-4} g), the model indicates that convection currents are reduced by a factor of 10^2, and predicts that particle losses will be lowered by more than a factor of 10, probably becoming negligible over the periods of time for which aerosols are likely to be stored in the ACPL (assumed to be 1-2 hours).

The aerosol considered consists of dry particles such as NaCl. It is assumed that the distribution of particle sizes is such that the cumulative spectrum of critical supersaturations is of the form $N = c\ S_c^{\ k}$, where k lies in the range 0.4 to 1.0.

It is also assumed, in conformicy with the specifications of aerosols to be used in the ACPL, that the concentration of particles with radii (r_p) exceeding 0.1 μm is negligible. The inconsistency between these two assumptions is of minor importance, since the estimates of depletion rates are necessarily rough ones, and the additional complexity of ensuring formal consistency by assuming a law of the form $N = c(S^k - S_o^k)$ would not be justified. The mechanisms considered are sedimentation, coagulation and convection-accentuated diffusion, as they affect the total count of CCN with $S_c < 10^{-2}$ (corresponding to particles with radii exceeding about 0.01 μm).

2. SEDIMENTATION LOSSES IN A STORED AEROSOL IN LOW GRAVITY

Since sedimentation depends directly on gravity, it is clear that in low gravity, the resulting depletion is likely to be extremely small. However, as pointed out in the Introduction, it is desirable to estimate the losses which would be expected to result from fall-out in 1-g, in order to check the models of depletion mechanisms against terrestrial experience.

Values of the mechanical mobility of aerosol particles (at 23C) have been listed by Fuchs (1964), as a function of radius. The particle radii of concern lie in the range from r_p = 0.01 to 0.1 μm. Over this range, the values of B may be fitted to an accuracy of better than 15% by the relationship:

$$B = 6.339 \times 10^{-2} r_p^{-1.787} \qquad (2.1)$$

where B is expressed in cm $sec^{-1} dyne^{-1}$, and r is in cm. (The error in this power law is in the sense of exaggerating B in the middle of the size range). Thus in this size range, the fall speed of a particle of radius r and density ρ_p is given approximately by:

$$V = 0.266\ \rho_p\ g\ r_p^{1.213}$$

Applying this to a well defined soluble aerosol such as NaCl, V may be expressed as a function of the critical supersaturation of the particle; in such aerosols,

$$r_p^3 = A\ S_c^{-2},$$

where

$$A = \frac{32\ M\ M_o^2 \sigma^3}{27R^3\ \rho_p\ \rho_o^2\ iT^3} \quad (=1.284\times10^{-22} \text{ for NaCl at 23C})$$

where M is the molecular weight of the aerosol material, M_o that of water, σ the surface tension, R the gas constant, ρ_p the density of the particles, ρ_o that of water and i the van't Hoff factor. Thus,

$$V = 0.266\ \rho_p\ g\ A^{0.404}\ S_c^{-0.809}$$

In the case of an aerosol with a cumulative distribution of particle critical supersaturations S_c of the form $N = cS_c^k$, S_c extending from S_1 to S_2, the flux of particles through a unit area of a horizontal plane is

$$0.266\ \rho_p\ g\ c\ k\ A^{0.404} \int_{S_1}^{S_2} S^{k-1.809}\ dS$$

Thus, the fractional loss per second due to sedimentation from a column of length L is:

$$\bar{E}_S = \frac{0.266\ \rho_p\ g\ k\ A^{0.404} (S_2^{k-8.809} - S_1^{k-0.809})}{L(k-0.809)\ (S_2^k - S_1^k)}$$

$$= \frac{0.266\ \rho_p\ g}{L} \left\{ \frac{k(r_2^{1.213-1.5k} - r_1^{1.213-1.5k})}{(k-0.809)\ (r_2^{-1.5k} - r_1^{-1.5k})} \right\}$$

where $r_2 (\approx 0.01\ \mu m)$ corresponds to S_2, and $r_1 (=0.1\ \mu m)$ to S_1.

The term in brace brackets depends on k, as shown approximately in the following table (1).

Table 1

k	Approximate values of $10^7 \bar{E}_S L/0.266\ \rho_p g$ (cgs)
0.4	2.136
0.6	1.828
0.8	1.520
1.0	1.379

In the case of a dry aerosol of NaCl (ρ_p = 2.16 g cm^{-3}) in terrestrial conditions, with L = 50 cm, $\bar{E}_S$ varies from $2.4 \times 10^{-6} sec^{-1}$ at k = 0.4 to $1.6 \times 10^{-6} sec^{-1}$ at k = 1.0, the loss rates per hour ranging from 0.9% to 0.6%. Thus, the contribution of sedimentation to particle loss in 1-g is not negligible, but as will be seen later, it is small compared with that due to diffusion in the presence of convection currents. In low gravity, the loss rates due to sedimentation are reduced by a factor of order 10^4, and are therefore quite negligible.

3. DEPLETION OF AN AEROSOL DUE TO COAGULATION

The effects of coagulation on the particle spectrum are complex, and depend on the initial size distribution. Some CCN will be eliminated by coagulation among themselves, and others will be formed by the coagulation of smaller particles. In an aerosol with a very large concentration of Aitken nuclei, coagulation may actually result in an initial increase in the concentration of CCN. However, in a more normal aerosol, as illustrated by the calculations carried out by Junge (1969), there is a decrease in the concentration of particles in the size range r_p = 0.01 μm to 0.1 μm; this however, is small compared with that which is usually observed to occur in containers of the size commonly used in terrestrial laboratories, or likely to be used in the ACPL. For the purposes of objective (2), as discussed in Section 1, it is therefore sufficient to derive an approximate upper estimate for the resulting loss rate.

This may be done in a simple manner by (1) ignoring the formation of particles with radii in the range $r_2 < r_p < r_1$, where r_2 = 0.01 μm and r_1 = 0.1 μm, as a result of the coagulation of particles for which $r_p < r_2$, and (2) using diffusion theory uncorrected for kinetic effects, which yields an overestimate of the rate at which particles within the range are eliminated by coagulation with each other.

Defining N in the usual way as the concentration of particles with critical supersaturations less than S_c and using the relationship $r^3 S_c^2 = A$, the concentration of particles in the range $r_p = r$ to $r + dr$ is

$$dN = -\frac{3}{2} ck A^{\frac{k}{2}} r^{-(1+\frac{3k}{2})} dr.$$

Thus, the rate at which coagulations occur per unit volume among particles within the range considered is:

$$\frac{9}{2} \pi c^2 k^2 A^k \int_{r_2}^{r_1} R^{-(1+\frac{3k}{2})} \int_{r_2}^{r_1} r^{-(1+\frac{3k}{2})} \{D(R)+D(r)\} (R+r) dr\, dR$$

Using equation 2.1, one may write

$$D(r)=kT\, B(r) = \alpha r^{\beta},$$

where $\alpha = 6.339 \times 10^{-2}$ $kT = 2.589 \times 10^{-15}$ (at 23C), and $\beta = -1.787$.

Substituting for D(R) and D(r) leads directly to the rate at which coagulations occur among particles in the range $r_1 > r_p > r_2$. The total concentration of such particles is

$$N = c A^{\frac{k}{2}} (\Delta r^{\frac{-3k}{2}}),$$

(Here, and below, (Δr^n) means $r_1^n - r_2^n$).

On substituting $c = NA^{\frac{-k}{2}} / (\Delta r^{\frac{-3k}{2}})$ and dividing by N, the fractional depletion rate is found to be:

$$\bar{E}_c = \frac{9\pi N k^2 \alpha}{\left(\Delta r^{-\frac{3k}{2}}\right) 2} \left\{ \frac{\left[\Delta r^{\beta-\frac{3k}{2}}\right]\left[\Delta r^{1-\frac{3k}{2}}\right]}{(\beta-\frac{3k}{2})\ (1-\frac{3k}{2})} \right.$$

$$\left. - \frac{2\left[\Delta r^{-\frac{3k}{2}}\right]\left[\Delta r^{1+\beta-\frac{3k}{2}}\right]}{3k\ (1+\ \beta-\frac{3k}{2})} \right\}$$

Putting $\beta = -1.787$, $r_1 = 10^{-5}$cm, $r_2 = 10^{-6}$cm, $(\bar{E}_c/N)$ may be calculated as a function of k. It is found that at k = 0.4, $\bar{E}_c = 7.9\text{x}10^{-10}$N, as a function of k. It is found that at k = 0.4, $\bar{E}_c = 7.9\text{x}10^{-10}$N, while at k = 1.0, $\bar{E}_c = 2.9\text{x}10^{-9}$N. For the aerosols to which the laboratory experience mentioned above applies, N is typically of order 10^3cm^{-3}; thus the depletion rates estimated as being due to coagulation range from 0.3% to 1% per hour.

Since kinetic effects, which result in decreased coagulation rates, have been neglected, and since the formation of particles within the range by the coagulation of smaller ones ($r_p < r_2$) has been ignored, these estimates are conservative. It may be concluded that, like sedimentation, coagulation cannot account for a major part of the depletion rates which are observed in terrestrial laboratories. In low gravity, loss rates due to coagulation will be the same as in 1-g; for $k < 1$, they should be somewhat less than (10^{-3} N) per cent per hour, where N is the concentration of particles in the range considered, i.e., 0.01 μm < r_p < 0.1 μm.

4. DIFFUSIONAL LOSSES IN A STORED AEROSOL IN 1-g AND LOW GRAVITY

A. Basis of Discussion

This discussion deals with the loss of particles with diffusivities (D) of order $10^{-5} cm^2 sec^{-1}$ (an average value for CCN), to the walls of a container with characteristic linear dimensions (L) of order 50 cm, over a period (t_s), of order one to two hours. Particle loss in low gravity (0.1 cm sec^{-2}) will be contrasted with that in terrestrial conditions.

The simplest approach is to consider the gas as being completely at rest. In that case, provided arrangements are made to withdraw samples from the center of the container rather than from a point close to a wall, the rate of loss of particles is very small indeed. For example, in the case of a sphere of radius a cm, after t sec , the central concentration of particles of diffusivity D $cm^2 sec^{-1}$ is reduced by the factor:

$$1 - \frac{2a}{(\pi Dt)^{1/2}} \sum_{n=o}^{\infty} \exp\left[-\frac{(2n+1)^2 a^2}{4Dt}\right]$$

Writing z for $\exp\left(\frac{-a^2}{4Dt}\right)$, since $z < 1$, the infinite series is dominated by the expansion of $\frac{z}{1-z^8}$. Hence, the proportional depletion is less than

$$\frac{2a}{(\pi Dt)^{1/2}} \quad \frac{z}{(1-z^8)}.$$

For $a = \frac{L}{2} = 25$ cm, $D = 10^{-5} cm^2 sec^{-1}$, $t = 3600$ sec., $\frac{a^2}{4Dt} \approx$ 4300, and the depletion is completely negligible.

If, on the other hand, it is supposed that the aerosol is thoroughly mixed just before a sample is removed, so that the air volumes which had been close to the walls are mixed with the relatively undepleted volumes near the center of the container, the rate of loss becomes significant. For example, the average concentration of particles in a sphere of radius a cm after t sec suffers a fractional depletion of:

$$\frac{6}{a}\left(\frac{Dt}{\pi}\right)^{1/2} + 3\frac{Dt}{a^2} - 12\frac{(Dt)^{1/2}}{a}\sum_{n=1}^{\infty} \text{ierfc}\ \frac{na}{(Dt)^{1/2}}$$

(e.g.Carslaw and Jaeger, p. 234).

The relevant values of $\frac{a}{(Dt)^{1/2}}$ are of order 10^2 (for $D = 10^{-5}$, $t = 3600$, $a = 25$). With such large values of the argument, it is convenient to write z for $\frac{na}{(Dt)^{1/2}}$, and to note that:

$$\text{ierfc}\ z = \frac{1}{\sqrt{\pi}} \exp(-z^2) - z\ \text{erfc}\ z\ .$$

Since ierfc z and erfc z are positive,

$$\text{ierfc}\ z < \frac{1}{\sqrt{\pi}} \exp(-z^2)$$

Hence the series of ierfc functions is dominated by the series

$\frac{1}{\sqrt{\pi}} \sum_{n=1}^{\infty} \exp\left(-\frac{n^2a^2}{Dt}\right)$, which itself is dominated by the geometric series

$$\frac{1}{\sqrt{\pi}} \sum_{n=1}^{\infty} \exp\left(-\frac{n\,a^2}{Dt}\right) = \frac{1}{\sqrt{\pi}} \frac{\exp\left(\frac{-a^2}{Dt}\right)}{\left\{1-\exp\left(\frac{-a^2}{Dt}\right)\right\}}$$

Since after one hour $\frac{a^2}{Dt}$ is of the order of 10^4, it is clear that the final terr in the expression for the fractional depletion is quite negligible; also, the second term is small compared with the first. Thus, the proportional reduction in concentration may be taken simply as $\frac{6}{a}\left(\frac{Dt}{\pi}\right)^{1/2}$; with a = 25 cm, D = 10^{-5}cm^{2}sec^{-1}, t = 3600 sec., this amounts to 0.026 (i.e. 2.6% in the first hour, the rate decreasing with time).

It is clear therefore that, if the air were completely at rest, it would be preferable not to stir the contents before taking a sample, However, there may be some motions present in the stored sample, and these could no doubt result in somewhat fluctuating sample concentrations as air volumes which have been close to the walls, and so have become depleted, drift randomly past the sampling point in the container. Stirring before sampling would then have the advantage of smoothing out these fluctuations, at the cost of a somewhat accentuated depletion rate.

Motions will be present in the stored sample because of

(a) The persistence of the initial stirring motions which must be generated when the sample is injected into the container.

(b) Thermal convection due to random temperature contrasts across the container, in the presence of a gravity field.

Some initial stirring may be essential in order to help ensure that the stored sample is homogeneous, despite any variations which may occur in the aerosol generator output; this stirring would then be an integral part of sample preparation. The stilling period required before the residual motions become negligible will depend on the manner in which the sample is injected into the container. Since this is not well defined, it will be assumed here that when a series of experiments are to be performed in the ACPL using an aerosol of only slowly varying properties, the first sample will be withdrawn only after a sufficiently long period has been allowed to elapse to ensure that residual motions have decayed to the point where their influence on particle depletion is negligible.

The following discussion of the persistent convective motions due to temperature contrasts envisages a container which is more or less like a cube of side L cm. The gravity vector is supposed to be arbitrarily oriented with respect to the walls, and the calculations are carried out assuming that the component parallel to each wall has a common value, appropriate to a randomly oriented direction, of $2\ g/\pi$. It is supposed that between opposite walls there exists a temperature contrast (δT) of order 0.1 C, and that a steady state convective circulation is established which will have the effect of continuously bringing fresh undepleted

air samples close to the walls (so increasing the overall rate of deposition of particles), and of sufficiently stirring the entire sample to keep it fairly well mixed.

The discussion of the convective circulation is relatively straightforward if conditions are such that the velocity boundary layer thickness is small compared with L, so that the convective flows along opposite walls are independent of each other, and if the time of transit of the air in the much thinner particle boundary layer along such a wall is small compared with t_s. It will be seen that such conditions probably exist in terrestrial laboratories, but not in the low gravity expected in the ACPL.

B. A Postulate Regarding Particle Depletion

Clearly, any investigation of particle depletion cannot be precise, since the shapes and sizes of containers in fact vary, and the nature of the random temperature gradients is unknown. However, given an approximate description of the convective flow field, it is possible to make an estimate of the rate of depletion in the presence of these motions. For this purpose, it will be hypothesized that when the fractional depletion is small, the loss of particles from the shearing flow close to the wall may be estimated using the plausible assumption that all particles are removed from the gas stream out to a distance y_1, where $y_1 = \sqrt{2Dt(y_1)}$, $t(y_1)$ being the time taken for the air to traverse the wall (of length L cm), completing this traverse to arrive finally at a point which is distant y_1 from the wall.

This postulate has been tested in two cases for which solutions are available.

(a) Flow in a tube

Gormly and Kennedy (1949) derived an asymptotic solution for the fractional depletion (f) which occurs in fully established laminar tube flow, which is valid when the depletion is small:

$$f = 2.56\ \mu^{2/3} - 1.2\ \mu - 0.177\ \mu^{4/3}$$

where $\mu = \frac{D\ell}{a^2\bar{u}}$ (ℓ = tube length, a = radius, $\bar{u}$ = mean velocity).

Using the postulate described above, the velocity at a distance y from the wall (y = a - r) is:

$$u = 2\bar{u}\ (\frac{2y}{a} - \frac{y^2}{a^2})$$

Hence the time of transit is:

$$t(y) = \frac{\ell}{2\bar{u}\ (\frac{2y}{a} - \frac{y^2}{a^2})}$$

Assuming $y_1 = \sqrt{2Dt(y_1)}$ and writing z for $\frac{y_1}{a}$,

$$z^3 - \frac{1}{2}\ z^4 - \alpha^3 = 0$$

where $\alpha^3 = \frac{D\ell}{2a^2\bar{u}}$

A first approximation to the solution of this equation is $z = \alpha$. Using Newton's method, the second approximation is

$$z = \frac{\alpha(1 - \frac{\alpha}{2})}{(1 - \frac{2}{3}\alpha)}$$

The flux of air passing through the tube out to radius r is $2\pi\bar{u}\ (r^2 - \frac{r^4}{2a^2})$. Hence, the fraction of the total flux which flows out between r = a and r = $(a - y_1)$, which is assumed to be totally depleted of particles, is

$$f = 4z^2 - 4z^3 + z^4$$

Substituting for z^4 from the original equation,

$$f = 4\ z^2 - 2\ z^3 - 2\ \alpha^3$$

$$= 4\ z^2\ (1 - \frac{z}{2}) - 2\ \alpha^3$$

Again, from (2), $(1 - \frac{z}{2}) = \frac{\alpha^3}{z^3}$, and therefore:

$$f = \frac{4\alpha^3}{z} - 2\ \alpha^3$$

Substituting the second approximation for z,

$$f = \frac{2\ \alpha^2\ (2 - \frac{7}{3}\ \alpha + \frac{\alpha^2}{2})}{(1 - \frac{\alpha}{2})}$$

$$\approx 4\ \alpha^2 - \frac{8}{3}\ \alpha^3 - \frac{\alpha^4}{3} + \ldots$$

Since $\mu = 2\alpha^3$, this reduces to:

$$f = 2.52\ \mu^{2/3} - 1.33\ \mu - 0.13\ \mu^{4/3} + \ldots$$

This expression will be referred to as the "second approximation".

In applying the postulate to estimate the diffusional losses in a stored aerosol, it is convenient, and appears quite adequate, to ignore the curvature of the velocity profile in the particle boundary layer, which is quite thin. In the case of tube flow, this simplification leads to the expression $u = \frac{4\bar{u}y}{a}$, and on solving $y_1 = \sqrt{2Dt(y_1)}$, it is found that $z = \alpha$. Using the same expression for u to calculate the flux of air in the layer y = 0 to $y = y_1$ leads to the result:

$$f = 4\alpha^2 - \frac{8}{3}\alpha^3$$

$$= 2.52\ \mu^{2/3} - 1.33\mu\ .$$

This expression will be referred to as the "first approximation."

When μ is small, so is f. At small values of μ, the first term in $\mu^{2/3}$ is dominant, and both approximations lie within 2% of the Gormly and Kennedy formula.

Table 2 compares the percent depletions predicted by the three formulae:

Table 2
f (per cent)

μ	From Gormly & Kennedy	First Approximation	Second Approximation	$z=\alpha$
10^{-3}	2.4	2.4	2.4	0.08
10^{-2}	10.7	10.4	10.3	0.17
10^{-1}	42.4	40.9	40.4	0.37

The final column shows values of z ($=y_1/a$), giving a measure of the relative thickness of the particle boundary layer within which depletion is significant (according to the first approximation).

(b) Flow in a (broad) channel

For the case of established laminar flow in a channel of depth 2 h, Kennedy (1953) has derived the asymptotic solution:

$$f = 1.1676\ \mu^{2/3} - 0.1\ \mu - 0.0175\ \mu^{4/3}$$

(where $\mu = D\ell/h^2\bar{u}$). This formula is applicable when μ (and f) are small.

Proceeding as before, the velocity of the air at a distance y from a wall is $\frac{3}{2}\ \bar{u}\ (\frac{2y}{h} - \frac{y^2}{h^2})$ so that the time of traverse is

$$t(y) = \frac{2\ell}{3\bar{u}\ (\frac{2y}{h} - \frac{y^2}{h^2})}$$

The postulate then leads to:

$$z^3 - \frac{z^4}{2} - \beta^3 = 0$$

where $z = \frac{y_1}{h}$ and $\beta^3 = \frac{2D\ell}{3\ h^2\bar{u}}$

As before, the first and second approximation to the solution for z are β and

$$\frac{\beta(1 - \frac{\beta}{2})}{(1 - \frac{2}{3}\ \beta)}\ ,\ \text{respectively.}$$

Of the total flux passing through the channel, the fraction lying within a distance y_1 from the two walls is equal to the fractional depletion:

$$f = \frac{3z^2}{2} \left(1 - \frac{z}{3}\right)$$

$$= \frac{3}{2} \beta^2 + \frac{1}{8} \beta^4 + \ldots$$

Since $\beta^3 = \frac{2}{3} \mu$,

$$f = 1.145\ \mu^{2/3} + 0.073\ \mu^{4/3} + \ldots$$

(the "second approximation")

In the case of channel flow, ignoring the curvature of the velocity profile near the wall leads to:

$$u = \frac{3\ y\ \bar{u}}{h}$$

and $$z = \frac{y_1}{h} = \beta$$

The same linear expression for u then leads to:

$$f = \frac{3}{2} z^2 = \frac{3}{2} \beta^2 = 1.145\ \mu^{2/3}$$

(the "first approximation")

Table 3 compares the percent depletion predicted by the three formulae:

Table 3

f (per cent)

μ	From Kennedy	First Approximation	Second Approximation	$z=\beta$
10^{-3}	1.16	1.15	1.15	0.09
10^{-2}	5.3	5.3	5.3	0.19
10^{-1}	24.1	24.7	25.0	0.41

As in Table 2, the final column gives a measure of the relative thickness of the particle boundary layer (according to the first approximation).

Tables 1 and 2 indicate that, provided the particle boundary layer is thin compared to the lateral dimension of the flow, the "first approximation" based on the postulate and taking u to be linear in y near the wall gives a useful estimate of the loss of particles from the shearing near-wall flow. As pointed out earlier, the use of the postulate in the following section to estimate depletion follows the method used in the "first approximation"; the additional complexity of proceeding to the "second approximation" does not seem warranted.

C. Independent Convective Flows Along the Walls

Assuming that such flows exist, a model can be used to determine a posteriori whether the thickness of the velocity boundary layer is indeed small compared with L, and the transit time in the particle boundary layer is small compared with t_s, as is necessary for the model to apply.

In this case, the flow may be approximated by the solution for convection in an infinite fluid (air) past a finite heated vertical plate which was given by Polhausen (e.g. Goldstein, p. 638). According to this solution, if x is the vertical coordinate along the wall and y is that normal to it, $\frac{u}{4\nu C^2 x^{1/2}}$ is a function of $C\, y\, x^{-1/4}$, where

$$C^4 = \frac{g(T_w - T_o)}{4\nu^2 T_o} ,$$

T_w being the wall temperature and T_o that of the fluid at infinity. The maximum value of u (u_m) occurs where $Cy\ x^{-1/4} \approx 1.0$. In terrestrial conditions ($g = \frac{2x980}{\pi}$) $cm^2 sec^{-1}$, taking $T_o = 293$ K, $T_w - T_o = \frac{\delta T}{2} = 0.05$ C and $\nu = 0.16\ cm^2 sec^{-1}$, $C \approx 1.1$. At a point halfway up one of the vertical walls, $x = \frac{L}{2}$ ($\approx$25 cm), so that the maximum velocity occurs where

$$y = \frac{1.0x^{1/4}}{C} \approx 2.2 \text{ cm}$$

(which is small compared with L).

The value of u_m at this point is about $0.27(4\nu C^2 x^{1/2}) \approx 0.9\ cm\ sec^{-1}$. At a distance of about 9 cm from the wall, the velocity (at x = 25 cm) has declined to about 0.08 cm sec^{-1}. Thus, in terrestrial conditions, the first condition of the model (velocity boundary layer thickness small compared with L) would appear to be met.

In considering the particle depletion which occurs in the presence of this flow, we are concerned only with a thin layer next to the wall, as will be seen later. Close to the wall, the relationship between

$$\frac{u}{4\nu C^2 x^{1/2}} \quad \text{and} \quad \frac{Cy}{x^{1/4}}$$

may be taken as linear (the first approximation method), so that:

$$u = 4\gamma\nu C^3 y x^{1/4}$$

where γ is a dimensionless constant ($\approx$0.6).

In the near-wall region, therefore,

$$\frac{\partial u}{\partial x} = \gamma\nu\ C^3 y x^{-3/4}$$

$$= -\frac{\partial v}{\partial y}\ , \text{ by continuity.}$$

Hence,

$$\frac{\partial v}{\partial y} = -\gamma \nu C^3 y x^{-3/4}$$

and $v = -\frac{1}{2}\gamma\nu C^3 y^2 x^{-3/4}$ (since v vanishes at y = 0)

Consequently,

$$\frac{Dy}{Dx} = \frac{v}{u} = -\frac{1}{8}\frac{y}{x},$$

so that a particle of air moves along a curve such that y varies as $x^{-\frac{1}{8}}$. This law obviously cannot apply as $x \to 0$, but the model used (an isolated finite vertical plate in an infinite fluid) cannot apply close to x = 0.

Thus a particle which finally reaches the top of the wall (where x = L) at distance y_1 from the wall has followed the path:

$$y = y_1 \left(\frac{L}{x}\right)^{1/8}$$

Substituting this value for y in the earlier expression for u gives an expression for u as a function of x only, for the particle which eventually passes through the point x = L, $y = y_1$:

$$u(x,y_1) = 4\gamma\nu C^3 L^{1/8} y_1 x^{1/8}$$

The time of transit of this particle along the wall is then:

$$t(y_1) = \int_0^L \frac{dx}{u(x,y_1)}$$

$$= \frac{2\ L^{3/4}}{7\gamma\nu C^3 y_1}$$

The particle depletion which occurs as the air moves along the wall is postulated to be that which would occur if all particles were lost from a layer of depth y_1, where

$$y_1 = \sqrt{2Dt(y_1)}$$

$$= \sqrt{\frac{4D\ L^{3/4}}{7\gamma\nu C^3 y_1}}$$

Thus,

$$y_1 = \left(\frac{4D}{7\gamma\nu C^3}\right)^{\frac{1}{3}} L^{\frac{1}{4}}$$

For terrestrial conditions ($g = \frac{2\times 980}{\pi}$ cm sec^{-2}), with $\nu = 0.16$ cm^2 sec^{-1}, $D = 10^{-5}$ cm^2 sec^{-1}, y_1 is about 0.1 cm. As discussed earlier, the maximum air velocity occurs at $y \approx 2.2$ cm at $x = L/2 = 25$ cm. Thus the "first approximation" discussed in paragraph B would seem quite applicable, since the particle boundary layer is so thin: the analog of z in Tables 2 and 3 is only about 0.05.

The time of transit along a wall at the limit of the particle boundary layer is given by:

$$t(y_1) = \frac{2\ L^{3/4}}{7\gamma\nu C^3 y_1}$$

$$= \frac{0.34\ L^{1/2}}{(\gamma\nu)^{2/3} C^2 D^{1/3}}$$

$$= 0.96\ \gamma^{-2/3} \left(\frac{\nu}{D}\right)^{1/3} \left(\frac{LT_o}{g\delta T}\right)^{1/2}$$

where $(T_w - T_o)$ is taken to be $\frac{1}{2}\,\delta T$.

In the terrestrial case ($g = \frac{2 \times 980}{\pi}$ cm sec^{-2}), with $D = 10^{-5}$ cm^2 sec^{-1} (an average value for CCN), $T_o = 293$, $\delta T = 0.1°C$ this amounts to about 520 sec., which is small compared with likely storage times, t_s of the order of one to two hours. Thus, the second condition of the model is also met.

Taking u to be linear in y in the particle boundary layer, the flux of air (F) per unit wall breadth reaching the top of the wall between y = o and y = y_1 is:

$$F = \frac{1}{2}\, y_1\, u\,(L, y_1)$$

$$= 2\gamma \nu C^3 L^{1/4} y_1^2$$

$$= 1.38\, \gamma^{1/3} \nu^{1/3} C\, L^{3/4} D^{2/3}$$

Substituting for C, with $(T_w - T_o) = \frac{1}{2}\,\delta T$,

$$F = 0.82\, \gamma^{1/3} \nu^{-1/6} \left(\frac{g \delta T}{T_o}\right)^{1/4} L^{3/4} D^{2/3}$$

$$= 0.69 \nu^{-1/6} \left(\frac{g \delta T}{T_o}\right)^{1/4} L^{3/4} D^{2/3}$$

in dimensional form.

Assuming that this flux of air has been denuded of particles as a result of diffusion to the wall, the resulting fractional loss of particles per wall per second is:

$$\varepsilon = 0.69 \nu^{-1/6} \left(\frac{g \delta T}{T_o}\right)^{1/4} n^{2/3} L^{-5/4} \quad \text{sec}^{-1}$$

for each wall which is ventilated by a convection current.

As discussed in Section 3, the diffusion coefficient D may be approximated by a power law, $D = 2.589 \times 10^{-15} r_p^{-1.787}$, in the size range 0.01 μm < r_p < 0.1 μm; further, in the case of a soluble aerosol such as NaCl, particle radius is related to critical supersaturation according to $r_p^3 = A/S_c^2$, where A (for NaCl at 23C) has the value 1.284×10^{-22} (r in cm, S_c in absolute units). Thus,

$$D = 2.589 \times 10^{-15} A^{-0.596} S_c^{1.191}$$

For an aerosol with a cumulative distribution of particles with critical supersaturation S_c of the form $N = cS_c^k$, where S_c extends over the range S_1 to S_2, the rate of particle loss per wall is

$$ck \int_{S_1}^{S_2} \varepsilon \, S_c^{k-1} \, dS_c \quad (cm^{-2} sec^{-1}).$$

Writing $\varepsilon = b D^{2/3}$, where

$$b = 0.69 \, \nu^{-1/6} \left(\frac{g \, \delta T}{T_o}\right)^{1/4} L^{-5/4} ,$$

$$\varepsilon S_c^{k-1} = 1.885 \times 10^{-10} A^{-0.397} b \, S_c^{(k-0.206)} .$$

Over the size range 0.1 μm < r_p < 0.1 μm, S_c, which varies as $r_p^{-3/2}$, varies by a factor of about 30. Consequently, provided k > 0.4, the fractional rate of loss $\bar{\varepsilon}$ of particles for which $S_1 < S_c < S_2$ is given by:

$$\bar{\varepsilon} = \frac{1.885 \times 10^{-10} A^{-0.397} bk \, S_2^{0.794}}{k + 0.794}$$

to within about 20%. For an NaCl aerosol, taking i = 2,

$\rho_p = 2.16$ g cm^{-3}, $A = 1.284 \times 10^{-22}$; for the environmental conditions discussed earlier ($\delta T = 0.1$C, $g = (\frac{2 \times 980}{\pi})$ cm sec^{-2}, $L = 50$ cm, $T_o = 296$ K), $b = 4.76 \times 10^{-3}$, and one finds:

$$\bar{\varepsilon} = 4.4 \times 10^{-4} \frac{k}{(k+0.794)} S_2^{0.794}.$$

For example, for $k = 1$, $S_2 = 10^{-2}$ (1 percent), $\bar{\varepsilon} = 6.3 \times 10^{-6}$. If as would seem to be likely, three of the six walls of the cube-like container are ventilated by convection currents, the air near the other three walls remaining at rest, the overall fractional depletion rate ($\bar{E}_d$) would be 1.89×10^{-5} sec^{-1}, and particles would be lost at a rate of 6.8% per hour from the whole stored sample, which is assumed to be well mixed. With $k = 0.4$, the loss rate is 4.2% per hour.

The analysis given above for terrestrial conditions would seem to indicate that convection currents caused by small temperature differences may be a major factor in causing depletion of an aerosol stored for a period of order 1-2 hours in a container with typical linear dimensions of order 50 cm. Since the fractional rate of loss is proportional to $\delta T^{1/4}$, even if $\delta T = 10^{-2}$C, the loss rate of particles up to $S_c = 10^{-2}$ is several percent per hour.

D. Application to Low Gravity

In the ACPL, where g will typically be reduced by a factor of 10^4, the rate of depletion by diffusional losses to the walls would appear from the above discussion to be reduced by a factor of 10. However, in this instance, the Polhausen solution predicts a velocity maximum some 20 cm from the wall, Clearly, the model does not apply; it is invalid to neglect the effect of the end walls, and in addition, the predicted opposing convective flows occurring on opposite walls would interfere with each other, since the thickness of the velocity boundary layer is comparable with L. It must be expected that in this case the Polhausen solution would seriously overestimate the convective velocities, and therefore could provide only an upper bound for the effects of convection on particle depletion.

Moreover, as shown above, the transit time of air along a wall at the limit of the particle boundary layer ($y = y_1$) varies as $g^{-1/2}$. Thus while the transit time in 1 g (some 500 sec) is small compared with t_s, that in low gravity is of order 5×10^4 sec (some 14 hours), which is large compared with t_s.

The analysis given above was based on the premise that the air stream which was depleted as a result of passing along the wall was mixed into the whole sample as a result of the general stirring caused by convection currents (which probably vary in time as the small, random temperature contrasts change). In low gravity, where the motion along the wall during the period t_s is only a

small fraction of the well length (L), and the maximum convective velocities are of order 10^{-2}cm sec^{-1}, this is clearly unrealistic. The anlysis therefore indicates that in low gravity, the most reasonable approximation would be simply to regard the air as being at rest. With a sampling point located in the center of the container, the diffusional loss rates would then appear to be negligible (once the initial stirring motions have been stilled).

5. SUMMARY AND CONCLUSIONS

Three mechanisms of depletion have been discussed, as they apply to a stored aerosol of dry particles of a soluble salt such as NaCl, the size distribution of which is such that it follows a law of the well known form $N = cS_c^{k}$. The fraction of the spectrum considered is that for which $S_c < 10^{-2}$ (1 percent), which corresponds to particles larger than about $r_p = 0.01$ μm. Particles larger than $r_p = 0.1$ μm are assumed to occur in negligible concentrations. The container is assumed to have linear dimensions of order 50 cm.

The models developed for the three mechanisms (sedimentation, coagulation and diffusion to the walls in the presence of convection currents) were checked by applying them to predict the depletion rates which would be expected in terrestrial (1-g) conditions. As shown in Table 4 below, sedimentation and coagulation (with an initial concentration of 10^3 particles cm^{-3}) appear to be minor causes of depletion compared with diffusion accentuated by convection currents. The total depletion rates predicted (of order 5-8% per hour) are in general agreement with laboratory experience.

Table 4

Estimated Rates of Depletion in 1-g of a Stored Aerosol (particles for which 0.01 μm < r_p < 0.1 μm)

Mechanism of Depletion	Depletion Rate (percent per hour)	
	k = 0.4	k = 1.0
Sedimentation	0.9	0.6
Coagulation ($10^3 cm^{-3}$)	<0.3	<1.0
Diffusion (w/convection present)	4.2	6.8
Total Depletion Rate	<5.4	<8.4

It may be concluded from the discussions of the three mechanisms that in low gravity in the ACPL, sedimentation losses will not be significant, nor will those due to diffusion, provided arrangements are made to withdraw samples from the center of the container rather than from near the walls. Depletion due to coagulation will depend on the aerosol, but, for the type of aerosol discussed, should be no more than $(10^{-3}N)$ % per hour, where N is the concentration of particles for which $0.01\ \mu m < r_p < 0.1\ \mu m$. (Coagulation is, of course, unaffected by gravity, but is strongly dependent on the initial size distribution - for example, if a sufficiently high concentration of Aitken particles [$r_p < 0.01\ \mu m$] is present, the concentration of particles in the size range considered may initially increase rather than decrease). In order to minimize diffusion-caused depletion, the container should be so designed that the contents are stirred as little as possible when samples are withdrawn from a central point.

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